

# RADIOACTIVITY IN THE AIR

## REFERENCES

- Aptec PCMCA/WIN manual.  
H. Enge, *Introduction to Nuclear Physics*, Chapt. 7.  
Evans, *The Atomic Nucleus*, pp. 518-522  
*Chart of the Nuclides*, G. E. -Knolls Atomic Power Laboratory.  
H.E. Johns & Cunningham, *The Physics of Radiology*, 3rd edition.  
W. R. Leo, *Techniques for Nuclear and Particle Physics Experiments*  
Segre, *Nuclei and Particles*, p. 156  
K. Siegbahn, *Alpha, Beta and Gamma-Ray Spectroscopy*, Vol. I, Chapt. 5, 8A.  
G.L. Squires, *Practical Physics*, pages 27, 202-204.  
G. N. Whyte and H. W. Taylor " A Radioactivity Experiment Using Activities Filtered from the Air" *American Journal of Physics* 30 120 (1962). (Available in room 229)

## INTRODUCTION

This experiment involves filtering outdoor air, and then measuring and identifying the radioactive substances deposited on the filter paper. The amount and type of radioactivity collected generally depend on the length of time for filtration, on the weather conditions on a given day and occasionally on emissions from industrial sources.

When this experiment was done in the early 1960s, the radioactive materials picked up on the filter paper were mostly fission products from nuclear bombs ( $^{90}\text{Sr}$ ,  $^{131}\text{I}$ , etc.), often in quite large amounts. In fact, there were networks of air and rain sampling stations around the world and the fallout from an atmospheric test was tracked as it drifted around the world. More recently, following the Chernobyl accident on 26 April 1986, air samples taken here in the Physics Department were found to contain  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$  and  $^{131}\text{I}$ . (These products arrived quite suddenly on 11 May 1986.) With the ban on *large atmospheric* tests being more or less successful and with the current general absence of major nuclear accidents, the radioactivity you measure is probably completely natural and has been around the environment for millennia.

The three natural radioactive families you are likely to observe originate with the isotopes  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{235}\text{U}$ . All three occur naturally with half-lives greater than the age of the earth. They each decay through a long chain of radioactive daughters, ending with stable  $^{206}\text{Pb}$ ,  $^{208}\text{Pb}$  and  $^{207}\text{Pb}$  respectively. The members of the chain should be determined from the Chart of the Nuclides posted in the lab, or from any number of texts (see the Evans or the Segre reference). In this experiment, the identification of the radioactive isotopes in your sample hinges on two criteria: the measured half-life and the type of radioactivity, alpha or beta. (Another possible means of identification could be by analysis of  $\gamma$ -ray energies. This method is an option suggested in the " $\gamma$ -Ray Spectra" experiment. )

$^{222}\text{Rn}$  (radon) is naturally present in the atmosphere because its progenitor,  $^{238}\text{U}$ , having a half-life of  $4.47 \times 10^9$  years (comparable to the age of the solar system) is naturally present in the earth.  $^{238}\text{U}$  decays by a succession of  $\alpha$  and  $\beta$  decays (the "(  $4n + 2$ ) series") to a chemically inert gas,

$^{222}\text{Rn}$ , which has a half-life of 3.82 days, long enough to diffuse out of the earth into the atmosphere. Once in the atmosphere, the  $^{222}\text{Rn}$  decays to chemically more active daughter products, starting with  $^{218}\text{Po}$ .

The results of your experiment measuring the  $\beta$ -decay curves, will depend in part on how long the filtration is carried out. What is actually being measured here is the radioactivity of dust particles, and in a long filtration the short half-life daughter products already deposited on the filter will decay. (See Figure 1.)

The rate at which  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$  (which are metallic atoms or ions) will accumulate on dust particles varies according to the availability of dust or smog. Thus in very clear conditions one might expect most of the  $^{218}\text{Po}$  to  $\alpha$  decay to  $^{214}\text{Pb}$  without first becoming attached to dust since the half-life is only a few minutes. There will then be relatively more accretion of  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  onto atmospheric dust. On the other hand, under very smoggy conditions there will be much more  $^{218}\text{Po}$  accretion on dust, and most of the observed radioactivity from filtered air may be decays from the filtered  $^{218}\text{Po}$ . The *proportions* of  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$  which are found initially on the filter may thus vary according to atmospheric conditions.

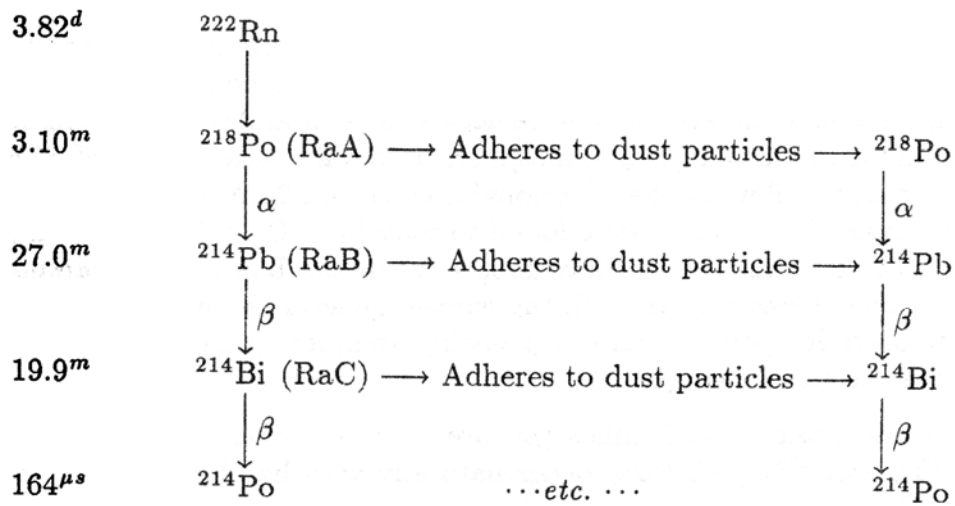


Fig. 1 - Model of successive radioactive decays from  $^{222}\text{Rn}$  as far as  $^{214}\text{Po}$ . The series terminates at  $^{206}\text{Pb}$  which is stable.

Another family of isotopes that you may detect comes from  $^{232}\text{Th}$  with a half-life of  $1.40 \times 10^{10}$  years. The "4n" succession of  $\alpha$  and  $\beta$  decays produces a shorter-lived (55.6 seconds) isotope of radon,  $^{220}\text{Rn}$  (Tn -thoron), which can decay in the air through various isotopes, including  $^{212}\text{Pb}$  (ThB),  $^{212}\text{Bi}$  (ThC),  $^{208}\text{Tl}$  (ThC"), ultimately ending up as stable  $^{208}\text{Pb}$ . Some of these isotopes will be detectable in your experiment, either from their  $\beta$  activity or from their  $\gamma$  emissions.

The "4n + 3" series originates with  $^{235}\text{U}$  ( $T_{1/2} = 7.04 \times 10^8$  years) and produces very small amounts of  $^{219}\text{Rn}$  (actinon) with half-life of 3.9 seconds.

In this experiment you have an opportunity to investigate the various radioisotopes in the air at the time of sampling and, in particular, to evaluate the concentration of radon in the atmosphere. Your chief technique will involve following the beta particle emissions of the isotopes collected on filter paper. You will produce a mathematical model for the decay of these isotopes, with several parameters to be defined. You will then fit these parameters to your data.

## THE EQUIPMENT

### Counting Equipment

**Geiger-Mueller Tubes:** These are gas filled ionization detectors with an applied high voltage large enough that electron and ion multiplication takes place, so that every time ionizing radiation passes through the counter and produces ionization, there is a measurable negative charge accumulated on the centre wire anode. With the anode connected through a resistor to the power supply, a standard voltage negative pulse appears at the anode (see W. R. Leo Chapter 6 or the Johns and Cunningham or the Enge reference).

**The Picker Scaler:** This provides a high voltage power supply for the G-M tube, in addition to being a counter.

Observe the construction of the G-M tube. Note the thin window at the end. *BE CAREFUL NOT TO DAMAGE IT.* This window has a thickness of between 1.4 and 2.0 mg/cm<sup>2</sup> which is thin enough to pass beta particles, but is thick enough to stop alpha particles of energy less than about 2 MeV. The gas is not very efficient for being ionized by  $\gamma$  rays so that this counter is most useful for beta particle detection.

To find out what kinds of pulses are produced by the GM tube, place a <sup>137</sup>Cs source in front of the GM counter, and connect it to the picker scaler "*detector input*" terminal with the high voltage set to a working value between 600 volts and 1,000 volts. Using the oscilloscope, observe the GM pulses appearing at the "*pulse input*" terminal. Note their voltage, shape and length. Note the effects of moving the source closer to and farther from the counter. Note the randomness of time of arrival of the pulses. Note the effects of varying the high voltage. Note the time the GM tube is "dead" after a pulse is produced. What is the "dead time" of this counter? Connect the oscilloscope to the "*output pulse*" terminal and observe the standardized pulse that the discriminator produces.

**CAUTION:** Some of the Picker scaler connectors have high voltages on them, sufficient to damage an oscilloscope input. Be extra careful in connecting the oscilloscope to the 'pulse input' terminal. Moreover, connect the oscilloscope to the Picker scaler 'pulse input' terminal only after the Picker scaler has been set into operation. Also, do not switch the picker scaler high voltage setting while the oscilloscope is connected. When in doubt, consult a demonstrator .

Determine the operating voltage of the GM tube by taking a plateau curve. With the <sup>137</sup>Cs as source near the front of the GM tube, measure and plot a curve of number of counts per standard time

interval versus high voltage. The almost-horizontal flat portion of this curve is called the plateau. The operating voltage should be taken in this region where counter sensitivity is independent of high voltage. While taking the plateau curve, be sure that you monitor the pulses on the oscilloscope to make sure that they 'make sense'.

NOTE: In doing this experiment, make sure that you count long enough at each voltage setting, in order to make sure that counting statistics allow adequate determination of the count rate. In drawing your plateau curve, include error bars, using as a guide the statement that the standard deviation in the number,  $N$ , of random events observed is equal to  $\sqrt{N}$ . See *W. R. Leo* chapters 1 and 4 or the *Johns and Cunningham* reference, page 541 to 545, or the *Squires* reference for material on counting statistics.

For this experiment you will accumulate your data using the **Aptec Multi-Channel Scaler (MCS)** function of the Multi-Channel Analyzer (MCA) card attached to a computer.

In the MCS mode, the analyzer acts as 4096 scalers which get switched on for successive time intervals or "*dwell times*". For this mode, standardized positive pulses of several volts pulse height are required. The analyzer will count the number of these pulses for the preset "dwell time" and store that number in channel 1, then count again for the dwell time, and store the new number in channel 2, etc. This mode is clearly useful for measurements such as half-life, where counts over successive time intervals are required.

To do multi-channel scaling of pulses produced by the GM tube certain arrangements must be made to provide the type of pulses required by the MCS input of the analyzer. To power the GM tube you will use a Picker scaler. The Picker scaler also serves the function of a discriminator, giving a more or less standard 3 volt negative pulse at its *pulse output* terminal for any GM pulse which is large enough. You can then feed this pulse into the input (in) connector of the pulse shaper. The output (out) of the pulse shaper a positive pulse which can be fed into the MCS input at the back of the analyzer. This positive pulse is compatible with the MCS input requirements.

Connect the GM tube to the Picker scaler, the pulse shaper, and the MCS (a 50 ohm termination on the pulse shaper output will often improve performance). Set an appropriate high voltage. Place a  $^{137}\text{Cs}$  as source near the detector. Follow pulses down the system, using an oscilloscope, to see what they look like and to check that they are satisfactory.

### **The MCARD software**

Open Program Manager → Aptec MCA. From Setup → ADC, you can setup the number of channels (the default setting uses all 4096). Confirm "Normal Spectrum" from Display. You will have to set the MCARD to "MCS Mode" (open Collect → Preset); set the "Dwell Time" (same menu), and set the Lower Level Discriminator (LLD) (open Collect → Settings)

*Note:* in order to erase the acquisition by the previous user, you may open File → Look at MCARD → Open → Refresh.

MCARDWin allows you to view MCARD properties and also to start, stop and clear data.

Note that the dwell time to be used for doing the count must be much less than the half-life being measured. It may also be worthwhile to check the activity of your sample 24 hours later to detect any long-lived isotopes. Over night data accumulation is recommended.

## Sampling

The air sampler can be found at the first year lab wicket in room 126. You will want to have it run between one and two hours. Your counting equipment should be ready to receive the sample when you turn off the sampler so that you minimize the time taken from when air sampling stops to when counting begins. It will be important to record the actual times of sampling and of data-taking.

## Approach to Data Analysis

There are various possible levels of sophistication in your approach to analyzing the data. On the most superficial level (worth, at most, one weight) you can merely do a constant background subtraction on your data and then, using a semi-logarithmic plot, find a rough half-life and total counting rate at the time sampling stopped. At the most complex level, you could do a full-blown analysis using the model described below, in addition to observing gamma-rays from the filter paper. You should read through the following pages and then decide for yourself the complexity of the analysis you wish to undertake.

## Processing Your Data and/or Calculate Your Model

The data collected on the Aptec MCPHA can be saved on a memory stick, or sent to your email as attachment and downloaded from there

This option allows you to import the Aptec data (all 4096 channels of it) into a Python script using the Poisson model.

## THE MODEL

Your model for counts registered in the detector consists of three parts:

1. The first part is the constant background observed when no sample is placed under the detector. This is the result of cosmic rays and gamma-rays present in the room. It is easily determined by making a sufficiently long background count.  
Since the amount of radioactivity in your sample is likely to be small, it is essential to determine the background (no sample) count rate accurately. Be sure to determine the background under identical conditions to those you will be using for your measurements, and count long enough to reduce the error in the background to  $\leq 1$  count per minute. As already indicated, if you observe  $n$  counts, the standard error is  $\sqrt{n}$ . (You can use the same multichannel scaler setup for observing background as you use in the decay observations.)
2. The second part is the decay of long-lived isotopes that are accumulated on the filter paper. These counts will probably be dominated by the decay of the "4n" series, generated by  $^{232}\text{Th}$ , which results in  $^{220}\text{Rn}$  (55.6<sup>s</sup>)  $\rightarrow$   $^{216}\text{Po}$  (0.145<sup>s</sup>)  $\rightarrow$   $^{212}\text{Pb}$  (10.64<sup>h</sup>)  $\rightarrow$   $^{212}\text{Bi}$  (1.00<sup>h</sup>)  $\rightarrow$   $^{212}\text{Po}$  (0.3<sup>μs</sup>) [alternative  $^{208}\text{Tl}$  (3.05<sup>m</sup>)]  $\rightarrow$   $^{208}\text{Pb}$ . The half-life of 10.64 hours of  $^{212}\text{Pb}$  will probably dominate this activity. However you may find other long-lived activities.  
These longer-lived components can be identified from a semi-logarithmic plot of the detector counts with background subtracted, for times after the half-hour activities from  $^{222}\text{Rn}$  have decayed. If a good linear fit is obtained, you can further subtract this component from the data. You now will have corrected data which will lead to the third part of the model.
3. The third part deals with the decay products of  $^{222}\text{Rn}$  and is detailed below.

## Secular Equilibrium

When radon ( $^{222}\text{Rn}$ ) alpha-decays, the resulting  $^{218}\text{Po}$  is highly ionized and some becomes attached to dust particles. (See figure I.) The subsequent beta decays produce  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  which are also highly ionized, have half-lives of the order of a half-hour and attach themselves to dust particles. The number of disintegrations per second of any isotope is proportional to the amount of that isotope. For example, the more  $^{218}\text{Po}$  there is, the more will decay per second and soon the amount of  $^{218}\text{Po}$  will build up to an equilibrium value which depends on the lifetime of  $^{218}\text{Po}$  and the amount of the  $^{218}\text{Po}$  being formed per second from its predecessor, radon. At equilibrium, the number of decays of  $^{218}\text{Po}$  per second is equal to the number of  $^{218}\text{Po}$  nuclei being created per second. But the number of  $^{218}\text{Po}$  nuclei being created per second must be equal to the number of radon decays per second and in the same way the number of  $^{214}\text{Pb}$  nuclei being created per second is equal to the number of  $^{218}\text{Po}$  decays per second. This continues down the chain and means that the number of decays per second of radon,  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ , and  $^{214}\text{Bi}$  are equal. This is known as secular equilibrium and holds whenever the local parent (in this case radon) has a long life compared to the subsequent generations. Thus, the measurement of the disintegration rate of anyone of the series will give the activity of the radon present. (See chapter 1 of the W. R. Leo reference for more on secular equilibrium).

Thus we should be able to measure the beta activity from dust in the air and account for it by considering only the decay of  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  if we eliminate alpha particles by using an aluminum filter thick enough to absorb all the alpha particles. *If this model fits the data*, then by determining the amount of one component (in our case  $^{214}\text{Pb}$ ) we can determine the concentration of radon in the atmosphere.

### The Mathematical Model for the Daughters of $^{222}\text{Rn}$

For a general treatment of radioactive decay chains, see chapter 1 of the W. R. Leo reference; for a detailed derivation of the following equations, see the *Whyte and Taylor* reference.

Although  $^{218}\text{Po}$ , being an alpha emitter, is not counted directly, its continuous arrival and rapid decay during the sampling period increases the amount of  $^{214}\text{Pb}$  present. However, once the filtering has stopped, the small number of  $^{218}\text{Po}$  atoms present has little effect (except for a small augmentation of  $^{214}\text{Pb}$  during the first 10 to 15 minutes) and the subsequent counting rate can be described solely in terms of  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$ . The following treatment will neglect any build-up of  $^{214}\text{Pb}$  from  $^{218}\text{Po}$  on the filter paper.

The variation of counting rate with time will be a function of the decay constants of  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  and of their relative activities at the start of the counting period. If we call  $A_B(t)$  = the actual activity of  $^{214}\text{Pb}$  (RaB) at time  $t$  and  $A_C(t)$  = the actual activity of  $^{214}\text{Bi}$  (RaC) at time  $t$ , measured from when air sampling is stopped. We consider a ratio  $R$  which is, at time  $t = 0$ . The relative amounts of isotopes "C" and "B" on the filter paper will be:

$$R = \frac{A_C(0)}{A_B(0)} \quad (1)$$

The actual observed counting rate in the detector is equal to the actual activity times a detector efficiency  $\varepsilon$  times a geometrical term. The detector efficiency is mainly the result of the beta particles of low energy being stopped by the aluminum foil alpha absorber and the GM tube window. Beta spectra are continuous from 0 to a maximum energy  $E_{\max}$ . As  $E_{\max}$  is considerably higher for  $^{214}\text{Bi}$  than for  $^{214}\text{Pb}$ , a smaller fraction of the beta particle spectrum is stopped by the foil and detector window in the former than in the latter, and thus  $\varepsilon$  is larger in the former. For an aluminum filter of 27  $\mu\text{m}$  thickness and a GM tube window of  $\rho x = 1.5 \text{ mg/cm}^2$ ,  $\varepsilon_C = 0.95$  for  $^{214}\text{Bi}$  (RaC) and  $\varepsilon_B = 0.80$  for  $^{214}\text{Pb}$  (RaB), giving  $\varepsilon_C/\varepsilon_B = 1.19$ .

Calling the actual observed activities:

$$A_B^{\text{obs}}(t) = \varepsilon_B A_B(t) \qquad A_C^{\text{obs}}(t) = \varepsilon_C A_C(t)$$

then at  $t = 0$ :

$$\frac{A_C^{\text{obs}}(0)}{A_B^{\text{obs}}(0)} = \frac{\varepsilon_C A_C(0)}{\varepsilon_B A_B(0)} = \frac{\varepsilon_C R}{\varepsilon_B}$$

Thus, the total count rate at  $t = 0$  is:

$$A_B^{\text{obs}}(0) + A_C^{\text{obs}}(0) = A_B^{\text{obs}}(0) \times \left( 1 + \frac{\varepsilon_C R}{\varepsilon_B} \right)$$

It is reasonably safe to assume that, because of the short lifetime of  $^{218}\text{Po}$ , very little gets attached to airborne particulates and that most of what does get onto the filter paper decays before the end of sampling. Thus we have decided to ignore any presence of  $^{218}\text{Po}$  on the filter paper.

You can derive the relations:

$$A_B^{\text{obs}}(t) = A_B^{\text{obs}}(0) e^{-\lambda_B t}$$

$$A_C^{\text{obs}}(t) = \frac{\varepsilon_C}{\varepsilon_B} A_B^{\text{obs}}(0) \left[ \frac{\lambda_B}{\lambda_C - \lambda_B} e^{-\lambda_B t} + \left( R - \frac{\lambda_B}{\lambda_C - \lambda_B} \right) e^{-\lambda_C t} \right]$$

where  $\lambda$  is the radioactive decay constant  $\lambda = \log_e 2 / (\text{half-life})$ .

The total observed counting rate as a function of time is:  $A_B^{\text{obs}}(t) + A_C^{\text{obs}}(t)$ .

### Comparison of the Model to Your Data

Part 3 of the model yields a predicted decay curve with two parameters,  $A_B^{\text{obs}}(0)$  and  $R$ . Your problem is to find if you can get a good fit and to establish the best values for the parameters. (This must be done on data which has first been corrected for parts 1 and 2.) A first approximation to  $A_B^{\text{obs}}(0)$  can be estimated by extrapolating the count rate curve back to  $t = 0$ , noting that total

counts at  $(t = 0)$  is:  $A_B^{obs}(0) + A_C^{obs}(0) = A_B^{obs}(0) \left( 1 + \frac{\epsilon_C}{\epsilon_B} \right)$ . A first approximation to  $R$  can be estimated, noting that the *Whyte and Taylor* reference calculates (using certain assumptions) a predicted value of  $R$  as a function of sampling time. Their plot of this relation is shown in Fig. 2.

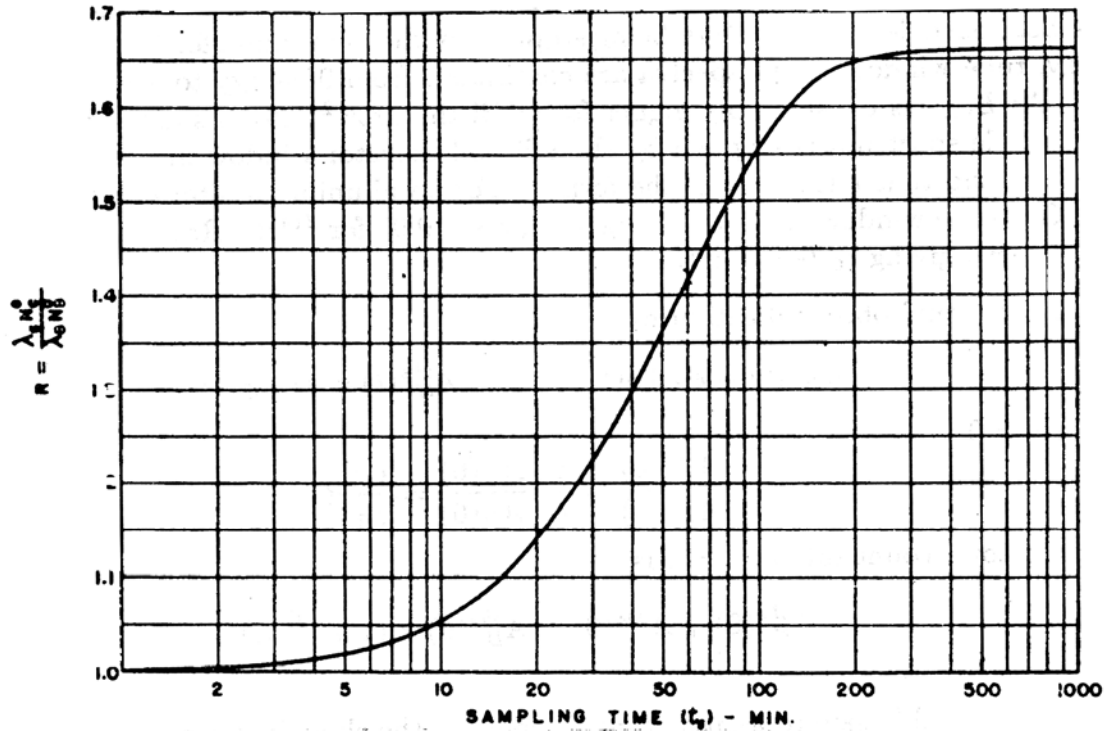


Fig 2. - Dependence of the ratio  $R$  on sampling time

One way of comparing your model to the data is by graphing data against model prediction, and using a least squares fit to a straight line through the origin. If you can minimize  $\chi$ -squared, and produce a reasonable value of  $\chi$ -squared, you probably have a good fit.

*Revised RMS 2009. Previous versions: dh- 1976, dalp, jp -1981, jbv -1990,1993.*